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Electrical and Optical Properties Dependence on Annealing Temperature for CdS Thin Films

KEYWORDS: CdS thin film, Thermal evaporation, Heat treatment, Optical band gap, Urbach energy, Electrical properties.

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ABSTRACT *Thin films of 100±10nm thickness CdS were deposited onto glass substrate maintained at room temperature using thermal evaporation technique. The effect of annealing temperature on the electrical and some optical properties were investigated. The annealing process in air for 1h was found to decrease the optical band gap energy and to increase the conductivity and Hall mobility of the investigated thin films. The optical band gap values were varied between 2.4 and 2.3eV with allowed direct transition. Urbach energy was found to be deceased for air-annealed films up to 623K then it increased with further elevation in annealing temperatures up to 723K. On annealing process, the dark DC electrical conductivity at room temperature and Hall mobility of the films increased from 1.67×10^{-6} to 3.93×10^{-4} $(\Omega.cm)^{-1}$ and from 0.31 to 42.35cm^2/V.s respectively .These physical behavior make prepared films good candidate for solar cells fabrication. All the investigated films have n-type conduction with their carrier concentration were to be in the order of 1013cm–3 .The activation energy as a function of the annealing temperature was found to be in the range 0.776-0.375eV.*

1. Introduction

Cadmium sulfide (CdS), a typical one of II-VI semiconductor materials, has received considerable interest for its useful applications in solar cells, photoconductors, sensors, a buffer layer on Cu (In, Ga) Se_2 (CIGS) thin films, optical detectors, field effect transistors, light emitting diodes and nonlinear integrated optical devices [1-9]. Owing to its transparency and photoconductivity, CdS thin films are also used widely as n-type window layers for thin films CdTe, $CuInSe_2$, Cu_2S and InP based heterojunction solar cells [1, 10-12]. Besides that, the wide optical direct band gap of CdS thin film (~2.4eV) is ideal to match solar spectrum for a photovoltaic window layer [13]. Particular properties achieved in films depend on the deposition method and the particular conditions of preparation [14]. Thus, several techniques have been employed to deposit CdS thin films of desirable optical, electrical and structural properties, such as thermal evaporation [15], chemical bath deposition (CBD) [16], spray pyrolysis [17], metal organic chemical vapor deposition (MOCVD) [18], close spaced sublimation [19], successive ionic layer adsorption and reaction (SILAR) [20], screen printing (SP) [21], electodeposition [22], RF-sputtering [23], pulsed laser deposition [24], electron beam evaporation [25], sol-gel [26],and ion layer gas reaction (ILGAR) [27],.

In the present work, thermal evaporation technique has been used to deposit thin films of CdS onto glass substrate at room temperature. The role of annealing temperature in air at 523, 623 and 723K on the electrical and some optical properties of CdS thin films were investigated.

2. Experimental procedure

CdS thin films were deposited at room substrate temperature (RT) onto cleaned glass slides by thermal evaporation technique using an Edward's high vacuum coating unit model E306A. During the evaporation, the vacuum was at 10^{-6} Torr. The source material was CdS powder with 99.999% purity from Balzer. The deposition rate was 0.5±0.05nm/s and the film thickness was about 100±10nm.The prepared samples were annealed at 523±10, 623±10, and 723±10K in air for one hour.

Optical absorption spectra were performed at room temperature using a Shimadzu UV-VIS spectrophotometer, UV-160 within the wavelength range 400-1100nm.

Electrical resistance of CdS thin films were measured under dark condition in the temperature range RT-473K using sensitive digital electrometer type Keithley 616. Hall voltage was measured as a function of current at constant magnetic field (0.257 Tesla) by using digital electrometer Keithley type 616.

3. Results and discussion

3.1. Optical properties

Study of materials by means of optical absorption provides a simple method for explaining some features concerning the band structure of materials [28].

Fig. 1 represents the optical absorption spectra (λ from 400 to 1100nm) for as-deposited CdS thin film grown on glass substrate and films annealed at 523±10, 623±10, and 723±10K for 1h in air. These measurement revealed that the absorbance increased with the decrease in wavelength, and sharp absorption edges were occurred at the wavelength λ<550nm. This behavior goes in line with result publishing by Elmas et al [29]. Another noticeable remark from Fig. (1) is that the absorption edge undergoes a shift towards the higher wavelength (lower photon energy) after annealing for 1h in air as a consequence of alteration in the optical band gap energy of these thin films.

From the optical absorption (A) data and the film thickness (t), the absorption coefficient (α) was calculated using the formula [30]:

$$\alpha = 2.303A/t$$

The nature of transition (direct or indirect) was determined according to Tauc relation [31]:

$$\alpha E=B(E-E_g)^r$$

where E is the photon energy, E_g is the optical band gap energy, B and r are constants and their values depend on the amorphousity of films. For allowed direct transition, r=1/2 and for allowed indirect transition, r=2.

Fig. (2) shows plots of $(\alpha E)^2$ versus E for as-deposited and annealed CdS thin films at 523, 623 and 723K. All the plots have shown straight line regions where $\alpha\geq10^4$ cm^{-1} indicates that CdS is a direct allowed band gap material.

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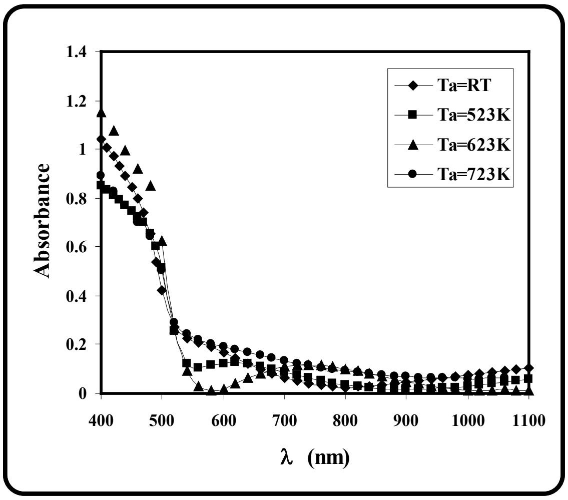
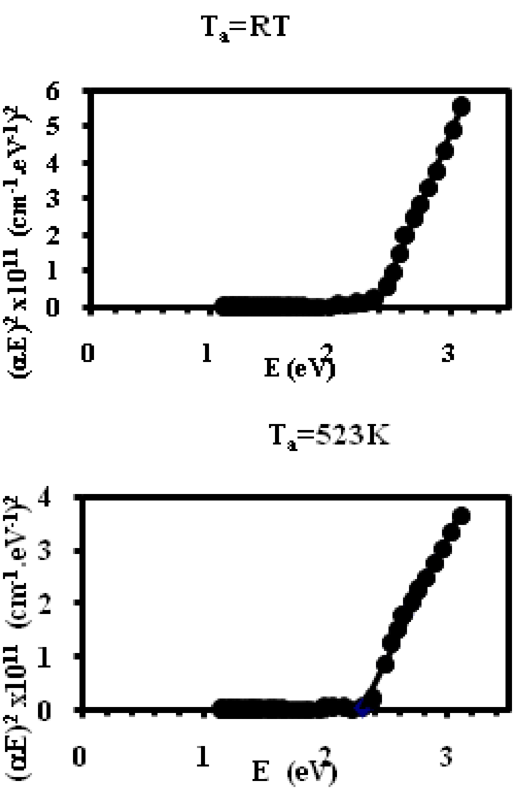


Fig. (1) Optical absorption spectra of as-deposited and air-annealed CdS thin films at different T_a.

The optical band gap energy has been estimated by extrapoling the linear portion of the absorption curves to $(\alpha E)^2$ axis . The band edge sharpness (B) value was obtained by taking the slope of the plots in Fig. (2) at the beginning of band-to-band absorption and is tabulated in Table 1. The value of B was observed to oscillate by thermal annealing and this behavior is in agreement with the literature [32] for CBD grown CdS thin films. They suggested that self-oxidation and sulfur evaporation were found to be responsible for this oscillating behavior. Also the plots in Fig. (2) exhibit exponential behavior in the lower photon energy range where $\alpha<10^4$ cm^{-1} and follow Urbach rule given by the expression [33]:

$\alpha = \alpha_0 \exp(E/E_u)$



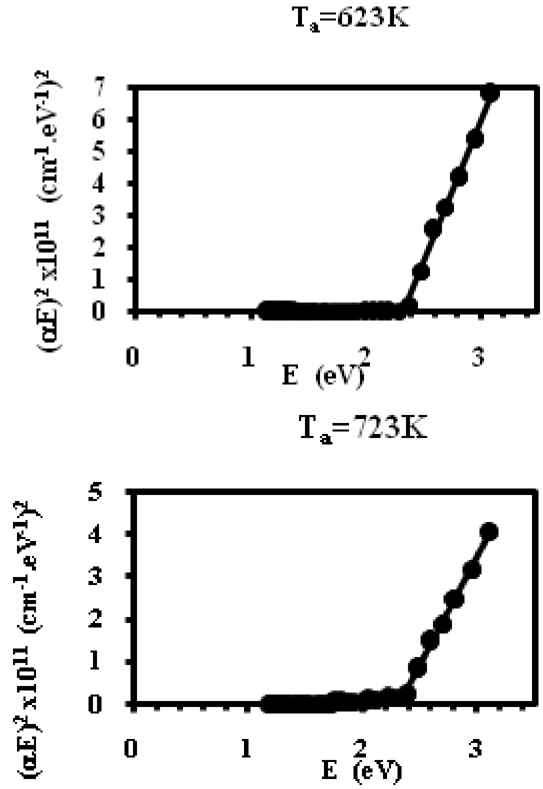
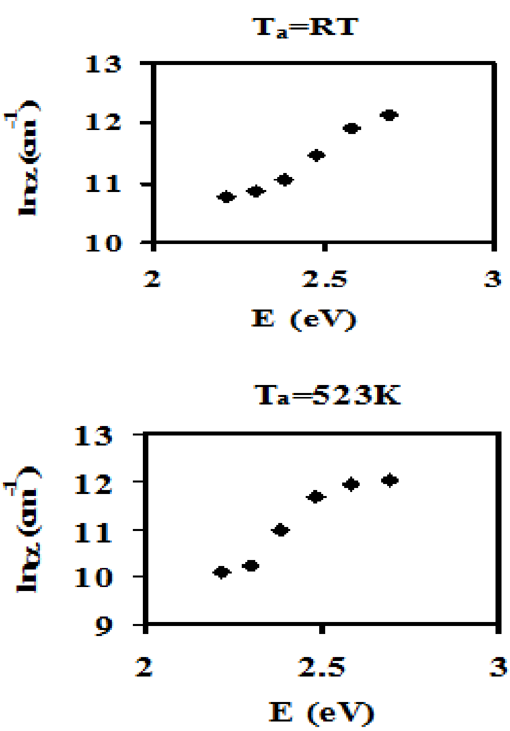


Fig. (2) $(\alpha E)^2$ vs E of as-deposited and air-annealed CdS thin films at different T_a..

where α_0 is constant and E_u is the Urbach energy, associated with the width of the tail and has been deduced from the inverse of the slope of lnα versus E plots and the graphs are presented in Fig. (3).



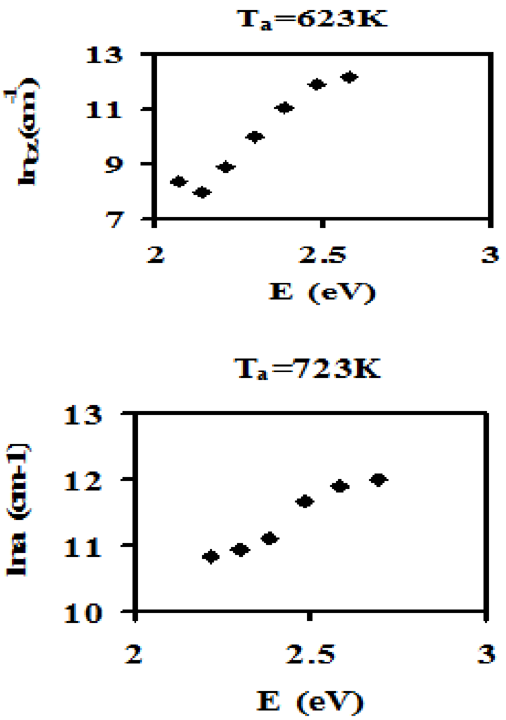


Fig. (3) Plots of lnα versus E of as-deposited and air-annealed CdS thin films at different T_a.

The dependence of the optical E_g and E_u of CdS thin films on T_a is shown in Figs. (4 and 5) respectively and it values are given in Table 1. It is clear that the annealed films shown a relative decrease in the optical E_g with annealing temperature. The optical band gap was found to be 2.4eV and 2.28eV for the as-deposited and the annealed films at 523±10K for 1h in air respectively. Then, a slight increase has been observed in the optical E_g with further increase of the annealing temperature up to 723±10K. The decrease in the values of E_g after annealing has been obtained by other researchers [26, 27, 32, 34-41], while Ramaiah et al [42] were found the direct band gaps of as-grown and annealed CBD deposited CdS films was 2.42 and 2.62eV with sub band gap of 2.35eV, respectively. Park [38] found that the band gap of the as-deposited CdS thin film grown by CBD technique was 2.83eV which is decreased to 2.53eV after annealing in a N_2 atmosphere for 1 h at 473K. According to his study, the band gap of the annealed CdS thin film increased and finally remained nearly constant at annealing temperature over 473K. It was suggested that the band gap changes with increasing annealing temperature due to changes in the defects, the compositions, the internal strain and the crystallite properties of the CdS thin films.

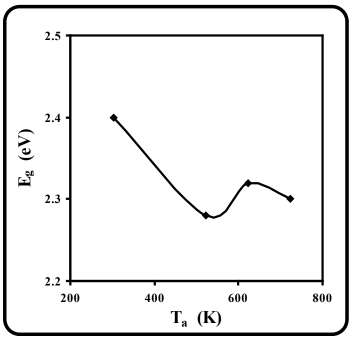


Fig. (4) Dependence of E_g on T_a for CdS thin films.

The Urbach energy of the as-deposited CdS thin film is 227.12meV and is reduced to 85.91meV with increasing annealing temperature to 623±10K as shown in Fig. (5) and Table 1. Further increase of T_a up to 723±10K resulted to an increase in E_u. A decrease in E_u value was also reported by Elmas et al [29] for ultrasonic spray pyrolysis CdS films. They found that E_u for as-deposited film was 133meV which then decreased to 98meV with increasing T_a up to 673±10K. The Urbach energy of prepared CdS thin films depends on the structural defects, dislocations density and some defects of the vacancy and interstitial states in the films [43]. Melin et al [32] conclude that the best annealing temperature for CBD grown CdS thin films was 673±10K and annealing over 673±10K was seen to degrade the optical properties of the film which was in agreement with our results for thermal evaporated CdS films.

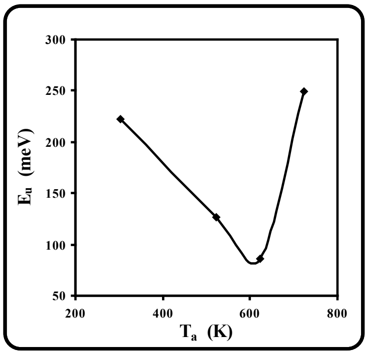


Fig. (5) Dependence of the E_u on T_a for CdS thin films.

3.2. Electrical Properties

The dark DC electrical conductivity (σ) of the as-deposited and annealed CdS thin films was measured in the temperature range RT-473K.

The dependence of the dark room temperature electrical conductivity (σ_{RT}) on annealing temperature of CdS thin films is shown in Fig. (6) and it values were tabulated in Table 2. It was observed that σ_{RT} for as-deposited CdS film was 1.67×10^{-6} $(\Omega.cm)^{-1}$ .It's value increased to 3.93×10^{-4} $(\Omega.cm)^{-1}$ after annealing CdS thin films up to 723±10K. This result indicated that annealed films to 723±10K showed higher σ_{RT} value by 2 orders compared to that of as-deposited film. An increase in σ_{RT} or a decrease in resistivity was observed by many authors [34, 35, 36, 40] .They attributed this behavior to the structural characteristics and impurity. Elmas et al [29] found that thermal annealing temperature up to 673±10K for 1h in air showed no significant effect on the electrical properties of the as-deposited CdS thin films grown by ultrasonic spray pyrolysis technique with thickness range 113-121nm.

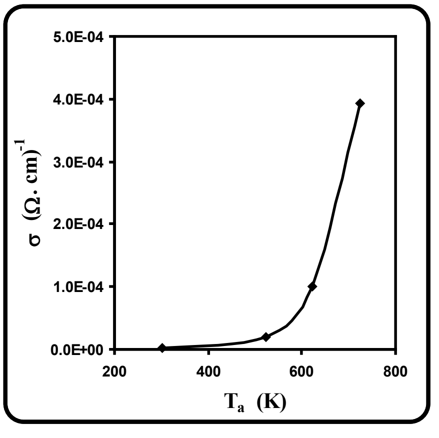


Fig. (6) Dependence of σ_{RT} on T_a for CdS thin films.

Table (1) Air annealing effects on values of E_g, B and E_u for CdS thin films.

T_a (K)	E_g (eV)	B x10^5 (cm^{-1} .$eV^{1/2}$)	E_u (meV)
As-dep.	2.40	8.94	227.12
523±10	2.28	7.07	126.88
623±10	2.32	9.48	085.91
723±10	2.30	7.07	249.02

Fig. (7)showed the variation of σ with temperature (T) in the form of lnσ versus 1000/T plots for as-deposited and annealed CdS thin films at 523±10, 623±10 and 723±10K for 1h in air. The conductivity of all the investigated thin films was increased with increasing temperature in the range RT-473K .This behavior is indicating that the films exhibited semiconducting manner. The plots of lnσ versus 1000/T were nonlinear showing two clear different regions. From the slopes of the curves in both regions, the activation energies (E_{a1}&E_{a2}) are calculated according to the following formula [43]:

$$\sigma = \sigma_0 \exp(-E_a/k_B T)$$

where E_a is the activation energy is absolute temperature and k_B is the Boltzmann's constant.

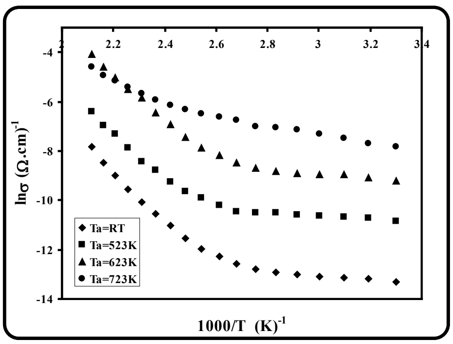


Fig. (7) Variation of lnσ with 1000/T of as-deposited and air-annealed CdS thin films at different T_a.

At relatively higher temperature (373-473)K, the conductivity was strongly dependent on temperature. Hence the conduction mechanism of E_{a2} in this case was due to carriers excited beyond the mobility edges into extended states. At relatively lower temperature (RT-373)K the conduction is due to carriers excited into localized states at the band edges [44].

Fig. (8) illustrated the dependence of the activation energies E_{a1} and E_{a2} on the annealing temperature for CdS thin films. From this figure and Table 2, the activation energy (E_{a2})

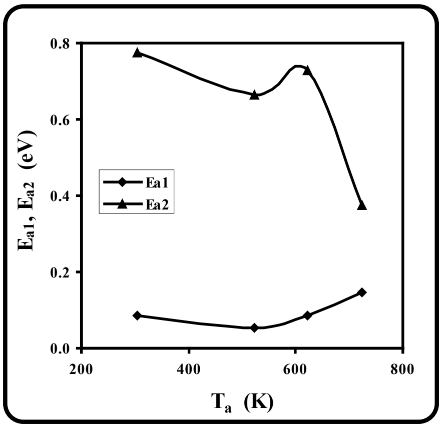


Fig. (8) Dependence of E_{a1} and E_{a2} on T_a for CdS thin films

for as-deposited CdS film is found to vary from 0.776eV to 0.375eV as T_a increased up to 723±10K. The behavior of the activation energy (E_{a1}) with annealing temperature was found to be increased as T_a increased from 523±10K to 723±10K. The values of E_{a2} were quite low compared to the value from the optical measurement (E_g/2). This result was also obtained by other researchers [29, 37, 41]. Metin et al [37] showed that the activation energy of CdS thin films grown by CBD technique and annealed in air from 373K to 773K was in the range 0.23-0.13eV in the low temperature region and 0.67-0.48eV in the high temperature region. They suggested that the low activation energy values obtained from the resistance measurements gives indication of doped levels (trapped levels or additional energy levels) due to the presence of impurity atoms in the forbidden gap of the semiconducting thin films. It is known that impurities and imperfections drastically affect the electrical properties of a semiconductor. In the same line of investigation the activation energy varies as 0.30-0.57eV for the lower temperature portion and 1.24-1.34eV for the high temperature portion were obtained by Devi et al [36] for as-deposited CdS thin film grown by CBD technique and annealed film at 523K in air for 1h.

By means of Hall measurements carried out at RT, carrier concentration and Hall mobility of the films have been calculated. Since the sign of the Hall coefficient was negative for all investigated films, indicating that electrons were predominant in the conduction process for all studied films.

Fig. (9) showed the variation of carrier concentrations (n) with different annealing temperature of CdS thin films. As shown in this figure and Table 2 the annealed CdS thin film at 523±10K was found to have a carrier concentration 7.2×$10^{13}$$cm^{-3}$ that's more than twice higher than that for the as-deposited thin film 3.4×$10^{13}$$cm^{-3}$. The increase of T_a up to 723±10K result an increase then decrease in values of carrier concentration. This result can be explained by the evaporation of sulphur from the films between 523 and 573K and by formation of CdO on film surface by oxidation at temperature above 573K [45].

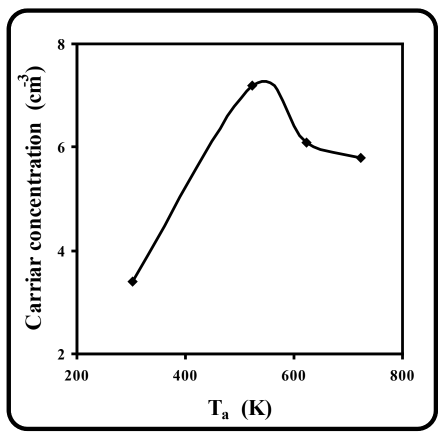


Fig. (9)Variation of carrier concentrations with T_a for CdS thin films.

The Hall mobility (μ_H) of the films was found according to the relation [46]

$$\mu_H = \sigma / ne$$

where e is the electronic charge.

The variation of the Hall mobility with annealing temperature for CdS thin films is shown in Fig. (10). From this figure and Table 2 μ_H value was observed to be increased from 0.306cm^2/V.s to 42.349cm^2/V.s for as-deposited to annealed CdS thin films up to 723±10K respectively. That is the charge mobility was increased by about more than two orders of magnitude. Similar behavior was observed by Bakiyaraj et al [40].

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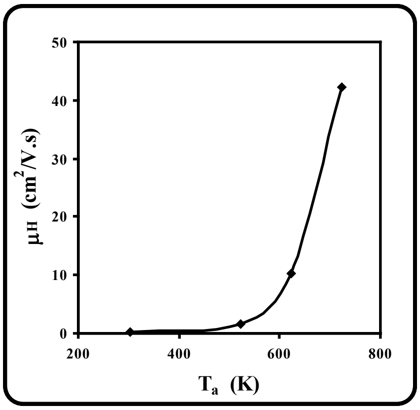

Fig. (10) Variation of μ_H with T_a for CdS thin films.

Table (2) Variation of σ, E_{a1}, E_{a2}, n and μH for CdS thin films with Ta .

T_a (K)	σ_{RT}x10^{-6} $(\Omega.cm)^{-1}$	E_{a1} (eV)	E_{a2} (eV)	nx10^{13} (cm^{-3})	μ_H (cm^2/V.s)
As-dep.	1.67	0.087	0.776	3.4	0.31
523±10	19.2	0.054	0.666	7.2	1.67
623±10	100	0.084	0.728	6.1	10.27
723±10	393	0.147	0.375	5.8	42.35

4. Conclusions

The effect of annealing temperature at 523±10, 623±10 and 723±10K in air for 1h on the electrical and some optical properties of thermal evaporated CdS thin films was investigated. From the results of the present work one can be concluded the following:

- Annealing in air resulted in a shift of the absorption edge towards higher wavelength and consequently a decrease in the optical band gap value.
- The minimum Urbach energy was observed in the vicinity of 85.91meV for film annealed at 623K.
- Dark DC electrical conductivity at room temperature increased by two orders of magnitude compared to that of as-deposited film after annealing up to 723±10K.
- The activation energy varied between 0.776 and 0.375eV with the increase of T_a.
- The carrier concentration was found to be in the order of $10^{13}cm^{-3}$.
- Hall mobility increased by about more than 2 orders of magnitude with increasing of the annealing temperature.
- The outcomes of the optimum values of physical properties for prepared films indicate it's well suited for solar cell fabrication.

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